

Magnetic structures in UCuSn

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We report neutron-diffraction results on UCuSn which crystallizes in the orthorhombic $P2_1cn$ structure. This compound undergoes two magnetic transitions around $T_N=62$ K and $T_M=25$ K. At $T=52$ K, UCuSn exhibits a noncollinear arrangement of uranium moments of $\sim 1.5\mu_B$ /U-atom with components in all three principal directions. Our diffraction data provide further evidence that the transition around 25 K is indeed magnetic in origin, although our refinements did not lead to a conclusive answer on its exact nature. © 2006 American Institute of Physics.

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The development of long-range magnetic order and the formation of complex magnetic structures in various uranium intermetallics have attracted the attention of many researchers in the past decades.¹ Among isostructural UTX (T , transition metal and X , p -electron element) compounds, the hexagonal GaGeLi-type ones have been reported to exhibit relatively localized behavior of its $5f$ electrons. UPdSn is the most extensively studied compound and it exhibits two magnetic transitions leading to complex noncollinear structures.^{2,3}

Here, we report on some recent neutron-diffraction studies on UCuSn with a focus on the magnetic transitions at low temperature. UCuSn was found to crystallize in the orthorhombic $P2_1cn$ structure, a structure closely related to the hexagonal GaGeLi structure.⁴ Szytula *et al.*⁵ proposed a hexagonal GaGeLi structure (space group, $P6_3mc$) for UCuSn and claimed that our proposed $P2_1cn$ crystal structure should be disregarded even though their model did not account for all observed Bragg peaks. Whereas a number of peaks remain unindexed assuming a hexagonal symmetry, our refinements at room temperature and 78 K result in high-quality fits of all the observed intensities with reduced χ^2 of, respectively, 3.88 and 3.81 and show no evidence for any secondary phase or off stoichiometry in our UCuSn sample.

Bulk studies on UCuSn reveal anomalies at about 25 and 62 K. We reported a sharp increase of electrical resistivity at around 62 K with lowering temperature, followed by a factor of 4 drop for temperatures below 25 K.⁴ In magnetization studies, in addition to anomalies in its temperature dependence at 25 and 60 K, there was some evidence of a small ferromagnetic component ($\sim 0.02\mu_B$ /U-atom) for temperatures below 25 K.⁴ The temperature dependence of the specific heat and the thermopower also revealed clear anomalies at those temperatures.⁵

In Ref. 4, we reported some preliminary neutron-diffraction studies on this compound. For temperatures below 60 K, we found additional magnetic contributions (see Fig. 1), all of which could be indexed in terms of the original orthorhombic crystallographic cell. There are magnetic contributions to the intensity at the positions of several nuclear peaks, but purely magnetic peaks appeared also at some crystallographically forbidden positions, such as (010), (100), and (101). In an attempt to determine the nature of the 25 K transition, we had also measured the temperature dependencies of a few selected peaks, but it did not reveal any sudden changes in the magnetic contributions at 25 K.⁴ However, the temperature studies at Chalk River Laboratories were severely hampered by the lack of neutron-beam intensity for some “decisive” high d -spacing magnetic reflections, such as the (010). It should be noted that our previous data are in good agreement with the data reported by Szytula *et al.*⁶ These authors reported an analysis of the observed magnetic

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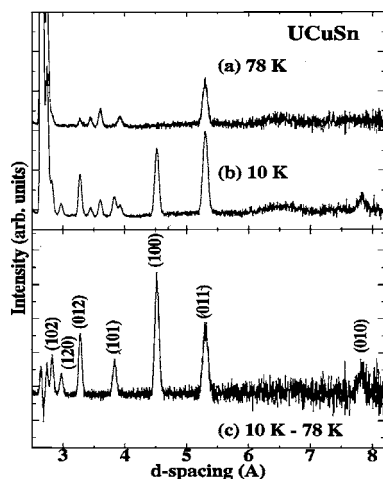


FIG. 1. Part of the raw data obtained on the HIPD at (a) 78 and (b) 10 K. The magnetic contributions, indexed in $P2_1cn$, are shown in (c) as the difference curve of data between 10 and 78 K. The intensities are normalized to the incident spectrum.

intensities for UCuSn by assuming an orthorhombic magnetic structure equivalent to the one reported for UPdSn for temperatures between 25 and 37 K.² Using the magnetic model proposed in Fig. 3 of Ref. 6, our attempts to calculate the magnetic intensities did not reproduce the values reported in Table I of that same reference. Furthermore, this model will not give any magnetic intensity on (010), which is observed experimentally (see Fig. 1).

Here, we report on recent results of additional neutron-diffraction experiments using the SPINS spectrometer at the NIST Research Reactor and some additional analysis of previous data taken on the high-intensity powder diffractometer (HIPD) at the Manuel Lujan, Jr. Neutron Scattering Center at Los Alamos. The sample used in the experiments at NIST was the same as used previously.⁴ We measured the temperature dependence of selected magnetic peaks using the SPINS spectrometer at the NIST Center for Neutron Research. We operated the SPINS spectrometer in a two-axis mode without the analyzer and a coarse collimation of open/80'/80'. The neutron beam was monochromated to a wavelength of 4.05 Å. We measured the Bragg intensities of five different reflections as a function of temperature. Mostly, we measured the peak intensities, but integrated intensities were measured about every 5–10 K by measuring about 20 points across each peak. In all cases, we found that the peak intensities scale very well with the integrated intensities (see Fig. 2). For all reflections, we find the onset of magnetic contributions below ~60 K, which is in good agreement with the

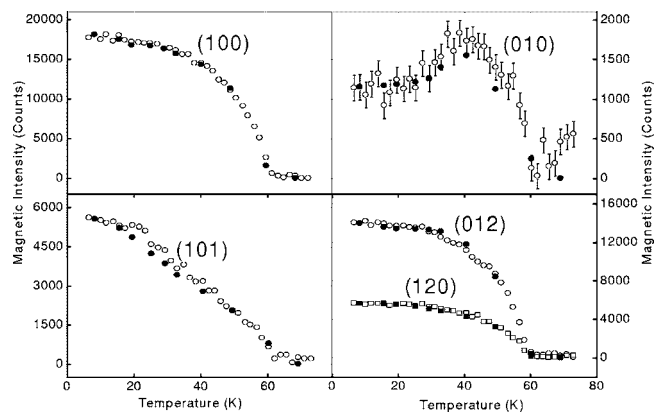


FIG. 2. Temperature dependencies of the magnetic peak intensities (open symbols) of sample Bragg reflections for UCuSn, as obtained on the SPINS spectrometer. The closed symbols represent the integrated intensities, normalized to the peak intensity value at 8.4 K. For both peak and integrated intensities, background and any potential nuclear contributions were subtracted, leaving purely magnetic contributions.

T_N reported from bulk investigations. Similar to the finding for UPdSn,³ we find a relatively smooth temperature dependence for most magnetic reflections with no indication of a second transition at 25 K. However, the (010) and (101) reflections exhibit anomalous behavior between 25 and 40 K. The drop in intensity for (010) and the irregular shape for (101) can be taken as an evidence for a second magnetic transition at lower temperatures (see Fig. 2).

The allowed magnetic structures for UCuSn can be derived from magnetic (Shubnikov) space group analysis, which generates four models, as shown in Fig. 3. It should be noted that all of the possible models allow for three moment

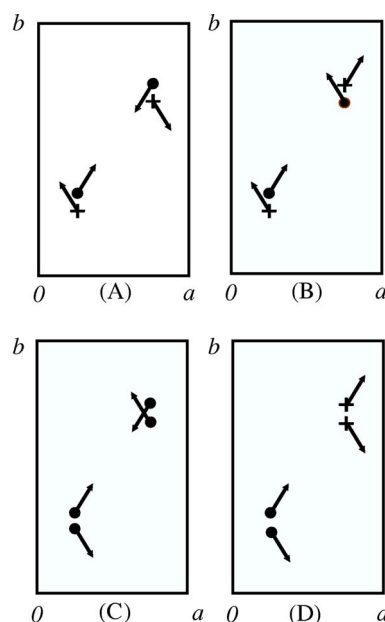


FIG. 3. The four possible magnetic structures (models A–D) for UCuSn based on magnetic space group analysis on $P2_1cn$, shown as projections on the orthorhombic a - b plane. The crosses indicate the z -axis components of the moments that point out of the plane, while the solid circles indicate the z -axis components that point into the plane. Our refinement indicates that the magnetic structure of UCuSn at 52 K is described best by the model A with magnetic parameters, as given in Table I.

TABLE I. Refined magnetic parameters of UCuSn at 52 K. The magnetic moment components are given with respect to the $P2_1cn$ notation.

$\mu_{Ux} = 0.74 \pm 0.01 \mu_B$
$\mu_{Uy} = 1.31 \pm 0.01 \mu_B$
$\mu_{Uz} = 1.31 \pm 0.02 \mu_B$
Total U moment = $1.54 \pm 0.01 \mu_B$
Reduced $\chi^2 = 3.41\%$
$wRp = 3.81\%$
$Rp = 2.48\%$

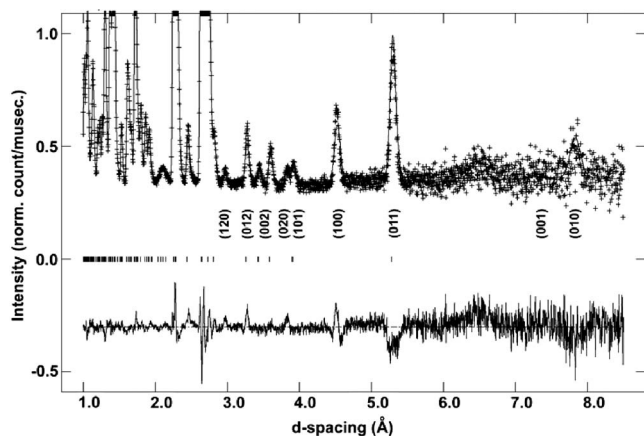


FIG. 4. Portion of the experimental diffraction pattern of UCuSn at 52 K in the $+40^\circ$ detector bank (symbols) and the fit to the data assuming model A in Fig. 3. The index markers indicate the position of the nuclear contributions, and some of the magnetic contributions, indexed in $P2_1cn$, are indicated. The intensities are normalized to the incident spectrum. At the bottom, the difference between the calculated and experimental intensities is given.

components; model A represents a noncollinear antiferromagnet and models B–D allow for a ferromagnetic component to be noncollinear antiferromagnetic. The existence of (011) and (101) reflections rules out any collinear magnetic moments linked by the $l=2n$ selection rule in $P2_1cn$. Moreover, the occurrence of the (010) reflection is evidence for a moment component out of the b - c plane. Finally, it should be noted that the (001) reflection is absent.

For the refinement of the nuclear and magnetic structures of UCuSn, we used the data obtained at 10, 25, 52, and 78 K using all six detector banks ($\pm 153^\circ$, $\pm 90^\circ$, and $\pm 40^\circ$) of the HIPD. We utilized the least-squares Rietveld refinement package GSAS.⁷ The different magnetic models were implemented as $Pna2_1$ (equivalent to $P2_1cn$ with interchanging the x and z axes) into the refinement. This allowed us to release the x position parameter for U, which had been arbitrarily fixed to define the origin for the crystallographic refinement.⁴ Assuming a single magnetic phase and the standard U^{3+} form factor,⁸ the antiferromagnetic model A is clearly preferred over the other three models for all temperatures below T_N (≈ 62 K). However, upon closer inspection of the magnetic intensities, we find that only the 52 K refinement is fully satisfactory under those conditions. A plot of the refinement is shown in Fig. 4 and the magnetic parameters at 52 K are given in Table I.

Similar refinements were attempted at 10 and 25 K, but we found that the above model generally overestimated the intensity at (010) while it underestimated the intensities of some peaks at smaller d spacings. In order to improve the low-temperature fits, we tried two different scenarios: (a) we assumed that there are two magnetic phases at low tempera-

tures and (b) a nonstandard uranium magnetic form factor. Scenario a was guided by the observation of a small ferromagnetic component in the magnetization below 25 K,⁴ which cannot be explained by model A. In this scenario we were able to achieve a reasonable fit of the 25 K data, which would imply that 95% of the sample exhibits model-A moment configuration and 5% of the sample exhibits model-D moment configuration (with a ferromagnetic net component along x). The refined moment value for this scenario is $1.903 \mu_B/\text{U-atom}$ ($\chi^2=3.33$, $wRp=3.74\%$, and $Rp=2.57\%$). Alternatively, scenario b was guided by the large drop in the resistivity at around 25 K (from almost semimetallic to metallic behavior),⁴ which is indicative of a “sudden” delocalization of the uranium $5f$ electrons. Thus, one might expect reduced orbital moments, similar to the case of URhAl.⁹ Taking the experimental magnetic form factor for uranium from Ref. 9, we were able to obtain a slightly worse (but still satisfactory) fit assuming model A only. In this case the refined value of U magnetic moment was $1.87 \mu_B/\text{U-atom}$ ($\chi^2=4.85$, $wRp=4.51\%$, and $Rp=3.05\%$). However, since neither model provides a complete agreement with the data it is possible that other alternatives should be considered.

In summary, we have shown that UCuSn exhibits two magnetic phases with transitions at about 62 and 25 K. While we are confident that both magnetic structures involve noncollinear arrangements of the uranium $5f$ moments, single crystals will be needed to unambiguously resolve the details of the moment configuration of UCuSn below 25 K.

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¹V. Sechovsky and L. Havela, in *Handbook of Magnetic Materials*, edited by K. H. J. Buschow (North-Holland, Amsterdam, 1998), Vol. 11.

²R. A. Robinson, A. C. Lawson, K. H. J. Buschow, F. R. de Boer, V. Sechovsky, and R. B. Von Dreele, *J. Magn. Mater.* **98**, 147 (1991).

³R. A. Robinson, A. C. Lawson, J. W. Lynn, and K. H. J. Buschow, *Phys. Rev. B* **45**, 2939 (1992).

⁴H. Nakotte, A. Purwanto, R. A. Robinson, K. Prokes, F. R. de Boer, L. Havela, V. Sechovsky, and I. P. Swainson, *J. Appl. Phys.* **79**, 6408 (1996).

⁵H. Kawanaka, H. Nakotte, E. Brück, K. Prokes, N. H. Kim-Ngan, T. Takabatake, H. Fujii, and J. Sakurai, *Physica B* **237–238**, 226 (1997).

⁶A. Szytula, M. Kolenda, V. H. Tran, R. Troc, F. Bourée, and R. Roisnel, *J. Magn. Mater.* **153**, L255 (1996).

⁷A. C. Larson and R. B. Von Dreele, Los Alamos National Laboratory Report No. LA-UR-86-748 (unpublished).

⁸S. W. Johnson, R. A. Robinson, H. Nakotte, E. Brück, F. R. de Boer, and A. C. Larson, *J. Appl. Phys.* **73**, 6072 (1993).

⁹J. A. Paixao, G. H. Lander, P. J. Brown, H. Nakotte, F. R. de Boer, and E. Brück, *J. Phys.: Condens. Matter* **4**, 829 (1992).